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13. ABSTRACT (Maximum 200 words) We have observed quasi-phase-matched second-harmonic generation in the reflection geometry from GaAs/AlAs multilayers. By using GaAs/AlGaAs multilayers as a cavity, we have also achieved cavity-enhanced nonphase-matched second-harmonic generation from GaAs/AlAs multilayers. The linewidth for the first-order reflection-second-harmonic generation is limited only by wave-vector mismatch. In addition, we have demonstrated two-order-of-magnitude enhancement on the conversion efficiency by using the cavity. Second, we have investigated effects of two-photon absorption on optical parametric oscillation. Third, we have measured spectra of two-photon absorption coefficient for CdSe and GaSe based on z-scan technique. Fourth, we have explored possibility of efficiently generating narrow-linewidth incoherent THz waves in multilayers based on transition radiation. Fifth, we have also proposed ultimate mechanism for Raman scattering of laser beam by single cyclotron electron in vacuum or cyclotron electrons in semiconductors. Sixth, we have grown and studied InP/InAs/InP quantum wires and GaAs/AlAs type-I and type-II superlattices. These structures can be eventually used for efficient infrared detection, Qswitched emitters, and optical communications. Finally, we have also achieved coherent blue light using cascaded quasi-phase-matched SHG and phase-matched SFG in partly-periodically poled KTP crystals with highest conversion efficiency of 3% (output power of 14 mW).					
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Final Report
Optoelectronics Devices Based on Novel Semiconductor Structures
(Transfer from Bowling Green State University to University of Arkansas)
Covered Period: June 1, 2000 – July 31, 2001

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Grant Number: F49620-00-1-0227

Status of effort:

We have made great progress in several areas. First of all, we have observed quasi-phase-matched second-harmonic generation in the reflection geometry from GaAs/AlAs multilayers. By using GaAs/AlGaAs multilayers as a cavity, we have also achieved cavity-enhanced nonphase-matched second-harmonic generation from GaAs/AlAs multilayers. The linewidth for the first-order reflection-second-harmonic generation is limited only by wave-vector mismatch. In addition, we have demonstrated two-order-of-magnitude enhancement on the conversion efficiency by using the cavity. Second, we have investigated effects of two-photon absorption on optical parametric oscillation. Third, we have measured spectra of two-photon absorption coefficient for CdSe and GaSe based on z-scan technique. Fourth, we have explored possibility of efficiently generating narrow-linewidth incoherent THz waves in multilayers based on transition radiation. Fifth, we have also proposed ultimate mechanism for Raman scattering of laser beam by single cyclotron electron in vacuum or cyclotron electrons in semiconductors. Sixth, we have grown and studied InP/InAs/InP quantum wires and GaAs/AlAs type-I and type-II superlattices. These structures can be eventually used for efficient infrared detection, Q-switched emitters, and optical communications. Finally, we have also achieved coherent blue light using cascaded quasi-phase-matched SHG and phase-matched SFG in partly-periodically-poled KTP crystals with highest conversion efficiency of 3% (output power of 14 mW).

Accomplishments/New Findings:

Below we concentrate on one of these projects, i.e. investigation of parametric processes by using transverse-pumping geometry.

GaAs and AlGaAs have very large second-order susceptibilities. To achieve efficient frequency conversion, multilayers have been used to achieve quasi-phase matching (QPM) [1]. There are two configurations for QPM: surface-emitting [1,2] and reflection [3-5]. In the surface-emitting geometry, a pump beam or second-harmonic beam propagates transversely or parallel to the surface normal while parametric or fundamental beams propagate in the layer plane. One of the advantages is the possibility of achieving oscillation without any cavity [2]. On the other hand, the reflection geometry can easily be used to measure nonlinearity of multilayers. In these two configurations, the dependence of the second-harmonic power on the propagation length is quite different. Although reflected-second-harmonic generation (SHG) in GaAs/Al_xGa_{1-x}As multilayers

was initially studied in Refs. [3-5], sharp QPM peak had not been achieved before due to (i) poor quality of the multilayers or (ii) lack of a tunable laser. In Ref. [3] 17 pairs of alternating layers of GaAs and $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ on a (110)-orientated GaAs substrate were used for forward and backward SHG (i.e. zero incident angle). *An enhancement by a factor of only 2.7 over the background was obtained because of the broad SH spectrum ($> 1000 \text{ \AA}$).* Moreover, part of the enhancement may be the result of Bragg condition, rather than QPM, since Bragg condition is too close to the broad SH peak. Ref. [4] illustrates how DBRs can enhance SHG for a *single thin layer* of $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ in a cavity grown on (100) direction. In this case, SHG was not quasi-phase-matched.

Recently, $\text{Al}_{0.8}\text{Ga}_{0.2}\text{As}/\text{GaAs}$ multilayers grown on a GaAs (100) substrate were used to demonstrate QPM at $1.064 \text{ }\mu\text{m}$ [5]. *However, the QPM peak (i.e. SH intensity vs. pump wavelength) was not directly measured.* The alternative measurement of enhancement vs. incident angle did not reveal a peak in the measurement range. The large enhancements were measured for the SH intensity relative to that for bulk GaAs. However, SH photon energy at $0.532 \text{ }\mu\text{m}$ is above the band-gaps of both GaAs and $\text{Al}_{0.8}\text{Ga}_{0.2}\text{As}$. In addition, since the multilayers can act as distributed Bragg reflection (DBR) due to a large difference of refractive indices, some of the enhancements may be attributed to the DBR rather than QPM.

We report our results on detailed investigation of reflection-SHG from GaAs/AlAs and GaAs/ $\text{Al}_{0.8}\text{Ga}_{0.2}\text{As}$ multilayers that have much higher quality. For the first time, we have directly observed a sharp QPM peak of the first-order reflection-SHG. An enhancement factor of about 124 over the background have been achieved. We have also observed QPM peaks at the second- and third-orders with and/or without a cavity. We have also measured the dependence of the SH power on the pump power. Furthermore, we have determined the relation among the SH polarization, SH power and pump polarization. Our investigation of the reflection-SHG from multilayers serves as a first step towards eventual applications of GaAs/AlGaAs multilayers for generating and amplifying mid-IR waves based on a novel configuration [6].

Our samples were grown using a Riber 32 molecular-beam epitaxy system at University of Arkansas. Fig. 1 shows their structure. 3-ps OPO pulses with a tunable output wavelength were used as a pump source. We first measured the spectra of the reflection-SHG for the samples. Fig. 2 represents a typical spectrum for an incident angle of 58° . To identify the origins for all these three peaks, we have also measured the reflection spectrum shown in Fig. 2. As one can see, two

peaks located at 1556 nm and 1699 nm, respectively, have the minimum reflectivities of the multilayer structure. Our calculations show that the two reflection dips at 1556 nm and 1699 nm are due to the Fabry-Perot cavity consisting of the sample surface (Fresnel reflection) and four-period GaAs/Al_{0.8}Ga_{0.2}As multilayers. Therefore, the two SH peaks at 1556 nm and 1699 nm are due to cavity-enhanced nonphase-matched SHG in GaAs/AlAs multilayers. On the other hand, there is no dip for the reflectivity of the multilayers at 1776 nm. Therefore, the peak in the reflection-SH spectrum located at 1776 nm (SH wavelength of about 888 nm) is due to QPM at the first-order in the GaAs/AlAs multilayers. At this wavelength an enhancement factor of about 124 over the background was demonstrated. This is the largest enhancement factor ever achieved solely using QPM multilayers. In addition, this is the first time ever to achieve such a large enhancement factor for the SH wavelength below the GaAs bandgap.

The linewidth of such a peak is only about 80 Å, which is much narrower than that obtained in Ref. [3] (over 1000 Å). In Ref. [3] the peak due to DBR is convoluted with the backward-SH peak. Therefore, it is difficult for us to estimate the linewidth of the backward-SH peak. In fact as shown in Ref. [3] the ratio of the backward and forward SH intensities exhibit a linewidth of over 1000 Å. This is due to the fact that owing to large fluctuations of thicknesses of the multilayers in the previous samples the linewidth is always limited by period fluctuations and other mechanisms as shown in Ref. [3]. Our measured linewidth is completely due to wave-vector mismatch (i.e. completely QPM reflection-SHG). The much narrower linewidth in our experiment shows that the quality of our multilayers is apparently much higher than that of our previous samples since our growth temperature is much more stable.

To confirm whether the second-order susceptibility used for QPM is originated from the GaAs layers, we have first measured the SH output powers for different polarizations of the pump beam. We have changed the polarization of the pump beam with respect to the x axis of the crystal by rotating the crystal around the surface normal. As one can see from Fig. 3, the SH power reaches a maximum value for a polarization angle of 45°. On the other hand, at the polarizations of 0° and 90°, the SH power reaches zero. We have observed that when the pump polarization is vertical, i.e. parallel to the crystal surface, the SH beam is always horizontal. These results obtained by us for the first time can be explained very easily. In fact, the element of the second-order susceptibility tensor used for QPM is d_{zy} . Only when the pump polarization is right between x and y axes of the crystal (45° with respect to both x and y axes), the SH power

can be maximized. In this case, the polarization of the SH beam must be a component of the SH polarization parallel to the z-axis of the crystal (surface normal). Thus, the SH beam is always horizontally-polarized. On the other hand, when the pump beam is polarized along the x axis or y axis of the crystal, the second-order nonlinear polarization can never be excited. Therefore, the SH power is zero. When the pump polarization is neither exactly between x and y axes nor parallel to the x axis or y axis, the polarization of the QPM SH beam is determined by a component of the SH polarization parallel to the z axis. As a result, the QPM SH polarization is always horizontal. In Fig. 3 we have shifted the measured polarization angles to use the x axis of the crystal as a reference. Such a result is different from Ref. [7] since only one pump beam is used.

We have measured the dependence of the SH output power on the average pump power at the first order (Fig. 4) using a power meter. We have confirmed the quadratic dependence for reflection-SHG. Such dependence has not been measured for the QPM multilayers. In a single AlGaAs layer with a cavity [5], however, power dependence was measured with a severe deviation from a quadratic dependence, attributed to two-photon absorption. In our measurements, however, we did not see any obvious deviation from the square law. At the pump wavelength of 1776 nm the highest average output power that we have achieved is about 13 nW (peak power of 57 μ W) for an average pump power of about 299 mW (peak intensity of about 25 MW/cm²). This corresponds to the conversion efficiency of 4.3×10^{-6} %. Since the propagation length for our structure is only about 2.5 μ m, one should compare the normalized conversion efficiency with those based on commonly-used materials such as BBO or KTP. The normalized highest conversion efficiency is about 69% cm⁻². On the other hand, the highest output power is about 26 nW at the pump wavelength of 1699 nm, which corresponds to a normalized conversion efficiency of 2.9×10^{-5} % /W.

Our next step in our research is to achieve difference-frequency generation and optical parametric amplification for generating and amplifying mid-IR beams (e.g. 980 nm + 1.55 μ m \Rightarrow 2.66 μ m while amplifying 1.55 μ m). We can then design, grow, and test an optimized structure for generating tunable and coherent waves in 3-5 μ m. Such a device can be used towards counter-measure and remote sensing. Such a device is compact, robust, and monolithic. Moreover, since the oscillation can occur without any feedback (mirror) output powers and wavelengths are ultrastable. On the other hand, if one uses a conventional and tabletop oscillator

for generating tunable 3-5 μm beam, a cavity is required. Therefore, output powers and frequencies are not stable. It is worth noting that it is not possible for semiconductor lasers to achieve such a large tuning range for the counter-measure and remote-sensing applications. Therefore, this project is crucial to one of the Air Force's missions: counter-measure. Reflection-SHG may be eventually developed to a technique for measuring indices of refraction. It can also be used to probe layer thickness and element concentration during the MBE growth of multilayers and to determine crystal orientation.

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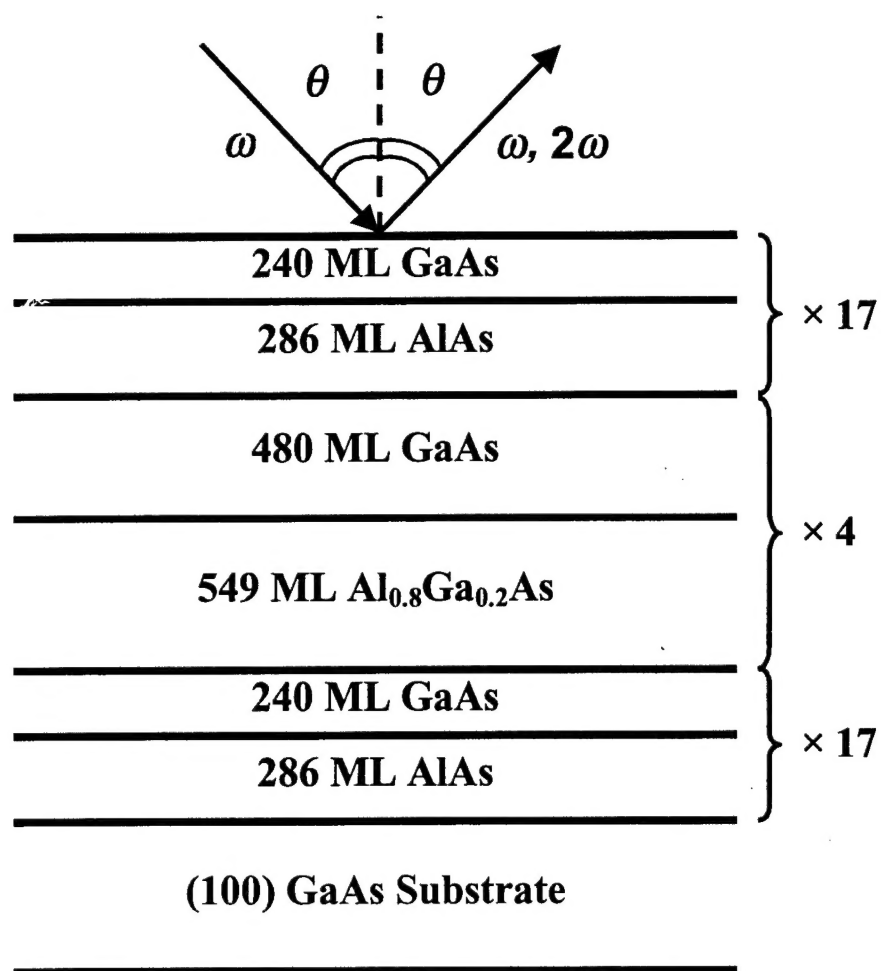


Fig. 1. Structure of GaAs/ $\text{Al}_{0.8}\text{Ga}_{0.2}\text{As}$ multilayers in the presence of a GaAs/AlAs multilayer cavity.

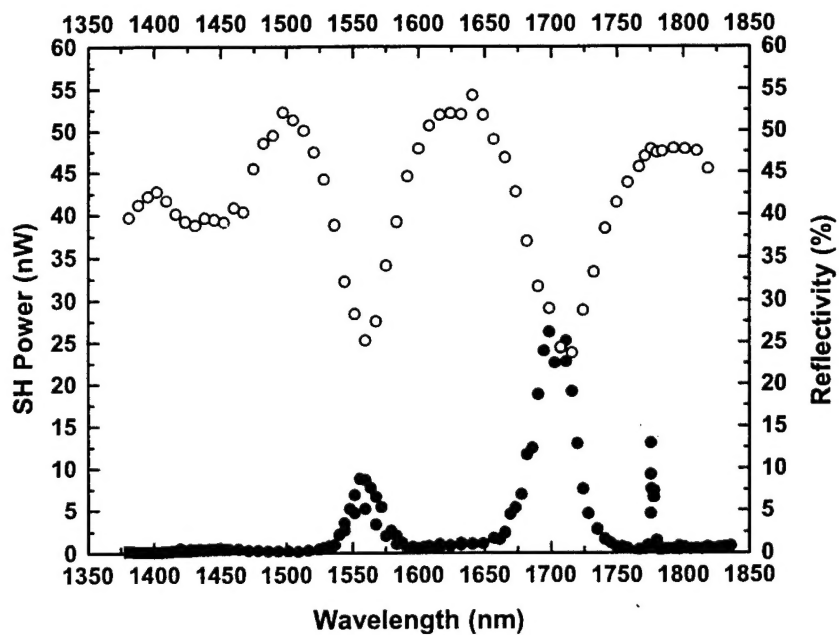


Fig. 2. Reflection-SH power vs. pump wavelength (filled circles) and reflectivity vs. incident wavelength (open circles) for an incident angle of 58° .

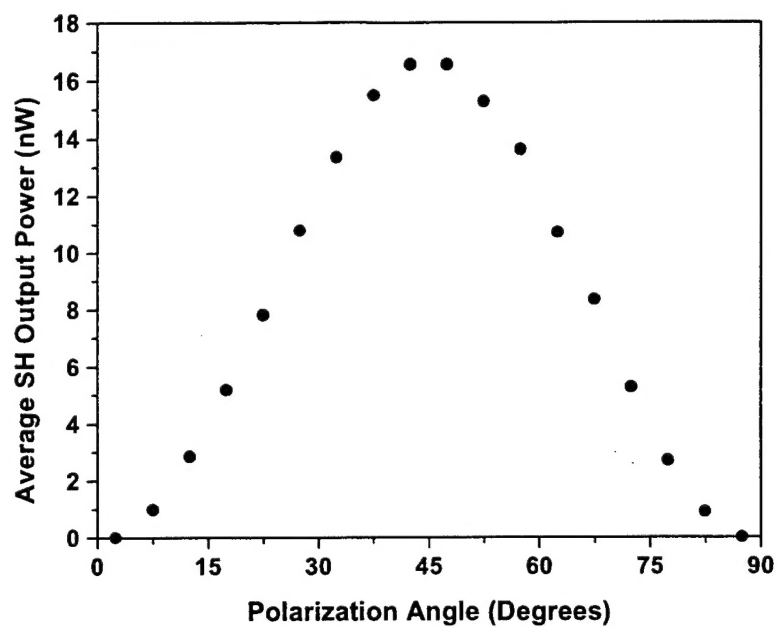


Fig. 3. SH power at the first QPM peak vs. polarization angle of the pump beam measured with respect to the x axis of the crystal. This was obtained by rotating the crystal around the surface normal while fixing the vertical pump polarization.

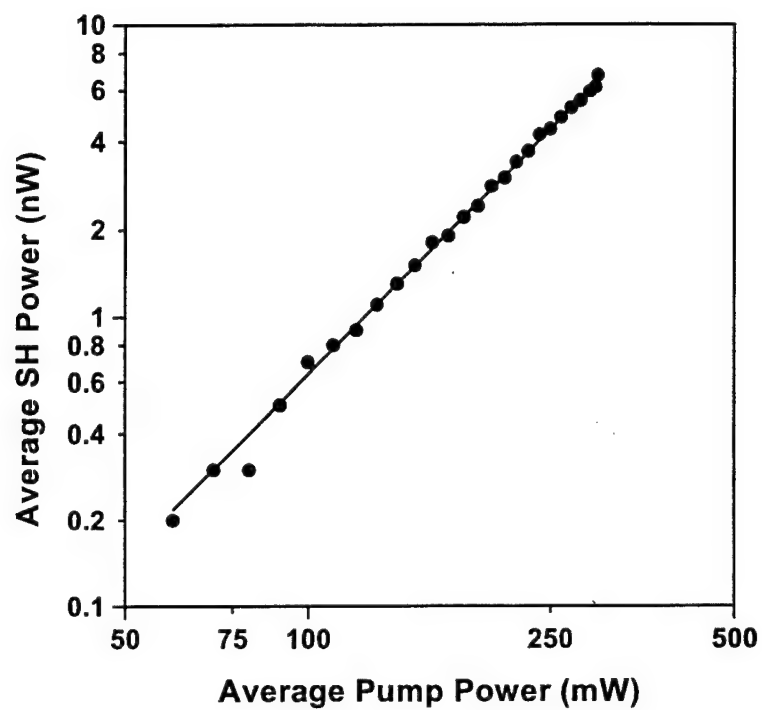


Fig. 4. Average SH power vs. average pump power at the QPM peak wavelength of 1776 nm (filled circles). The fitting (solid line) clearly shows a quadratic dependence.

Personnel Supported:

Y. J. Ding (faculty); I. B. Zotova and X. Mu (students).

Publications

X. Mu, I. B. Zotova, Y. J. Ding, H. Yang, and G. J. Salamo, "First observation of anomalously large blue shift of photoluminescence peak and evidence of band-gap renormalization in InP/InAs/InP quantum wires," *Appl. Phys. Lett.*, vol. 79, pp. 1091-1093, Aug.20, 2001.

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W. Shi, Y. J. Ding, X. Mu, X. Yin, and C. Fang, "Electro-optic and electro-mechanical properties of poled polymer thin films," *Appl. Phys. Lett.*, vol. 79, pp. 3749-3751, Dec. 3, 2001.

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Interactions/Transitions:

[(*) - refereed papers; () - invited papers]**

* W. Shi, Y. J. Ding, Xiaodong Mu, and N. Fernelius, "Tunable and coherent radiation in the range of 2.7–29 μm based on phase-matched difference-frequency generation in GaSe," CLEO'02.

* I. B. Zotova, X. Mu, and Y. J. Ding, and J. B. Khurgin "Reductions of threshold for mid-IR optical parametric oscillation by intracavity optical amplifier," CLEO'02.

* Wei Shi and Yujie J. Ding, Nils Fernelius, and Konstantin Vodopyanov, "Coherent and widely-tunable THz and millimeter source: new application for GaSe," CLEO'02.

* Y. J. Ding, "Tunable THz oscillators based on synchronously-pumped optical parametric oscillation in multiple-bonded, orientation-patterned, and single GaAs plates," QELS'02.

* X. Mu, Y. J. Ding, H. Yang, and G. J. Salamo, "Vertically stacking 10 periods of self-assembled InAs/InP quantum wires," CLEO'02.

* X. Mu and Y. J. Ding, "Third- and second-harmonic generation from 1.319- μm Nd:YAG laser in one KTiOPO₄ crystal," CLEO'02.

* X. Mu and Y. J. Ding, J. B. Khurgin, X. Wang, J. Zhang and F. Choa, "Growth and characterization of GaAs/AlAs superlattices: evidence of quasi-indirect transition between minibands," CLEO'02.

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X. Mu and Y. J. Ding, J. B. Khurgin, X. Wang, J. Zhang and F.-S. Choa, "Design, growth, and characterization of GaAs/AlAs type-II superlattices," Quantum Dot Sources and Detectors, Optoelectronics, Photonics West'02, San Jose, CA, Jan. 20-25, 2002.

** Y. J. Ding, "Forward and backward harmonic generation in periodically-poled LiNbO₃ and KTiOPO₄ crystals," OSA Annual Meeting, Oct. 14-18, 2001, Long Beach, CA.

X. Mu and Y. J. Ding, "Coherent all-solid-state blue source based on frequency-tripling in partly-periodically-poled KTP crystal," OSA Annual Meeting, Oct. 14-18, 2001, Long Beach, CA.

X. Mu, I. B. Zotova, Y. J. Ding, H. Yang, and G. J. Salamo, "Band-filling effect and band-gap renormalization in self-assembled InP/InAs/InP quantum wires," OSA Annual Meeting, Oct. 14-18, 2001, Long Beach, CA.

H. Abu-Safe, X. Mu, and Y. J. Ding, "Tunable coherent mid-IR source based on difference-frequency generation in periodically-poled multi-grating LiNbO₃," OSA Annual Meeting, Oct. 14-18, 2001, Long Beach, CA.

** Y. J. Ding, "Novel approaches to generation, amplification and detection of THz waves," 5th Mediterranean workshop and topical meeting, "Novel optical materials and applications", May 20 -27, 2001, Cetraro, Italy.

* Y. J. Ding, "New mechanism for efficient generation of narrow-linewidth THz beams in multilayers and superlattices pumped by electron beam," QELS'01, May 6-11, 2001, Baltimore, MD.

* X. Mu, Y. J. Ding, H. Yang, and G. J. Salamo, "First-order quasi-phase-matched reflection-second-harmonic generation from GaAs/AlAs multilayers," CLEO'01, May 6-11, 2001, Baltimore, MD.

* I. B. Zotova and Y. J. Ding, "Spectrum of two-photon absorption coefficient for GaSe," CLEO'01, May 6-11, 2001, Baltimore, MD.

* X. Mu and Y. J. Ding, "Efficient generation of red and blue light from 1.32 μ m in partly-periodically-poled KTiOPO₄ crystal," CLEO'01, May 6-11, 2001, Baltimore, MD.

* Y. J. Ding, "Raman scattering of laser beam by single cyclotron electron: ultimate mechanism," QELS'01, May 6-11, 2001, Baltimore, MD.

** J. B. Khurgin and Y. J. Ding, "Semiconductor quantum-dot optical amplifiers," Ultrafast Phenomena in Semiconductors V, Photon. West, Jan. 20-26, 2001, San Jose, CA.

Y. J. Ding and I. B. Zotova, "Role of Two-photon absorption in optical parametric oscillation and amplification," Ultrafast Phenomena in Semiconductors V, Photon. West, Jan. 20-26, 2001, San Jose, CA.

X. Mu and Y. J. Ding, "Reflection-second-harmonic generation from semiconductor multilayers," Ultrafast Phenomena in Semiconductors V, Photon. West, Jan. 20-26, 2001, San Jose, CA.

** Y. J. Ding, "Efficient generation of coherent blue and green light in KTiOPO_4 crystals," LEOS Ann. Meet. Nov. 2000.

X. Mu and Y. J. Ding, "Efficient generation of blue light based on second-harmonic generation and sum-frequency generation in periodically-poled KTP crystal," TuJ2, OSA Ann. Meet. 2000.

I. B. Zotova and Y. J. Ding, "Effect of two-photon absorption on optical parametric oscillation," TuJ1, OSA Ann. Meet. 2000.

G. Beadie, W. S. Rabonovich, Y. J. Ding and X. Mu, "Transversely-pumped, transversely quasi-phasematched, integrated parametric oscillator," ME1, OSA Ann. Meet. 2000.

* Y. J. Ding, "Efficient generation of narrow-linewidth THz waves from multilayers pumped by electron beam," IQEC 2000, QThD0055, Sep. 10-15, 2000, Nice, France.

Patent disclosures:

Y. J. Ding and X. Mu, "Coherent blue-light generation in a single partly-periodically-poled potassium titanyl phosphate crystal" (Provisional).

Y. J. Ding and W. Shi, "A new widely-tunable and coherent terahertz source implemented by using a gallium selenide crystal" (Pending).

Honors/Awards:

Topical Editor for J. Opt. Soc. Am. B. (2001-present).

Member of QELS 2002 Program Committee for Nonlinear Optics.

Member of program committee for SPIE Photonic West'01 and '02 Int. Sym. on "Ultrafast Phenomena in Semiconductors V and VI".